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Uranium isotopes in well water samples as drinking sources in some settlements around the Semipalatinsk Nuclear Test Site, Kazakhstan

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Abstract Radiochemical results of U isotopes (^{234}U , ^{235}U and ^{238}U) and their activity ratios are reported for well waters as local sources of drinking waters collected from the ten settlements around the Semipalatinsk Nuclear Test Site (SNTS), Kazakhstan. The results show that ^{238}U varies widely from 3.6 to 356 mBq/L (0.3–28.7 $\mu\text{g/L}$), with a factor of about 100. The ^{238}U concentrations in some water samples from Dolon, Tailan, Sarzhal and Karaul settlements are comparable to or higher than the World Health Organization's restrictive proposed guideline of 15 μg (U)/L. The $^{234}\text{U}/^{238}\text{U}$ activity ratios in the measured water samples are higher than 1, and vary between 1.1 and 7.9, being

mostly from 1.5 to 3. The measured $^{235}\text{U}/^{238}\text{U}$ activity ratios are around 0.046, indicating that U in these well waters is of natural origin. It is probable that the elevated concentration of ^{238}U found in some settlements around the SNTS is not due to the close-in fallout from nuclear explosions at the SNTS, but rather to the intensive weathering of rocks including U there. The calculated effective doses to adults resulting from consumption of the investigated waters are in the range 1.0–18.7 $\mu\text{Sv/y}$. Those doses are lower than WHO and IAEA reference value (100 $\mu\text{Sv/y}$) for drinking water.

Keywords Semipalatinsk nuclear test site · Kazakhstan · Well waters · Uranium isotopes · Annual effective dose

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Introduction

Over a period of 40 years from 1949 to 1989, the former Union of Soviet Socialistic Republics (USSR) conducted more than 450 nuclear explosions at the Semipalatinsk Nuclear Test Site (SNTS), Kazakhstan; 26 of them were above ground, 87 in the atmosphere and 346 underground [1, 2]. Considerable efforts have been devoted to investigate the consequences of radiation exposures to the residents living in the area, particularly in villages contaminated heavily by fallout of the radioactive cloud [1–3].

We have also investigated the present situation of radioecology in and around the SNTS since 1994, and measured long-lived radionuclides ^{137}Cs and Pu isotopes in large number of soil samples from various areas [4–7]. From these measurements, settlements around the SNTS we visited were found to be contaminated by $^{239,240}\text{Pu}$ with levels from several to a few hundred times higher than those (40–120 Bq/m²) for global fallout observed in Japan, while ^{137}Cs contamination is not so high [8]. Furthermore,

as for an external radiation dose in the air, it has been gradually clarified that residents of Dolon, where is well known to have been highly contaminated by radioactive fallout due to the first USSR nuclear detonation on 29 August 1949, received around 0.5 Gy [9].

On the other hand, information concerning internal doses experienced by village residents is still very limited around the SNTS. Recently, Tanaka et al. [10] reported that frequencies of unstable-type chromosome aberrations and micronucleus in lymphocytes were higher in residents of contaminated areas such as Dolon, Sarzhal and Kainar than those of the non-contaminated area. They point out that such a higher incidence may be caused mainly by internal exposure, although factors such as age, habitation, smoking, drinking water, medical exposure, life style and so on must be further considered in the interpretation of data from contaminated area.

To serve as an aid to resolve such problem, the present work was aimed at clarifying the present situation of radionuclide levels in well waters as local sources of drinking waters. Among naturally occurring radionuclides, uranium belongs to the most chemical and radiological toxicity of elements for human. Here, we report the present U isotope (^{234}U , ^{235}U and ^{238}U) levels in well waters collected mainly from the settlements around the SNTS and the associated annual effective doses to adults resulting from consumption of the investigated waters.

Experimental

Samples

The settlements where well waters were collected are shown in Fig. 1. These areas are semiarid plains with a low mean annual precipitation (200–300 mm). Total of 35 well water samples was collected from the contaminated settlements such as Dolon, Sarzhal and Karaul around the SNTS. The pH measurement in the water was carried out on a potable pH meter (Model-D-24, Horiba Ltd.) that was calibrated in situ before each set of measurement. The samples were taken in two 200 mL polyethylene bottles without filtration. In addition, about 100 mL of water was collected in light-tight glass bottle for measuring alkalinity; diluted mercuric chloride solution was added to the bottle to prevent decomposition of dissolved organic matter and then the bottle was tightly sealed. For comparison, river surface water sample was also collected from the Irtysh River, which is the largest one among them and flows into the Ob River after leaving this area.

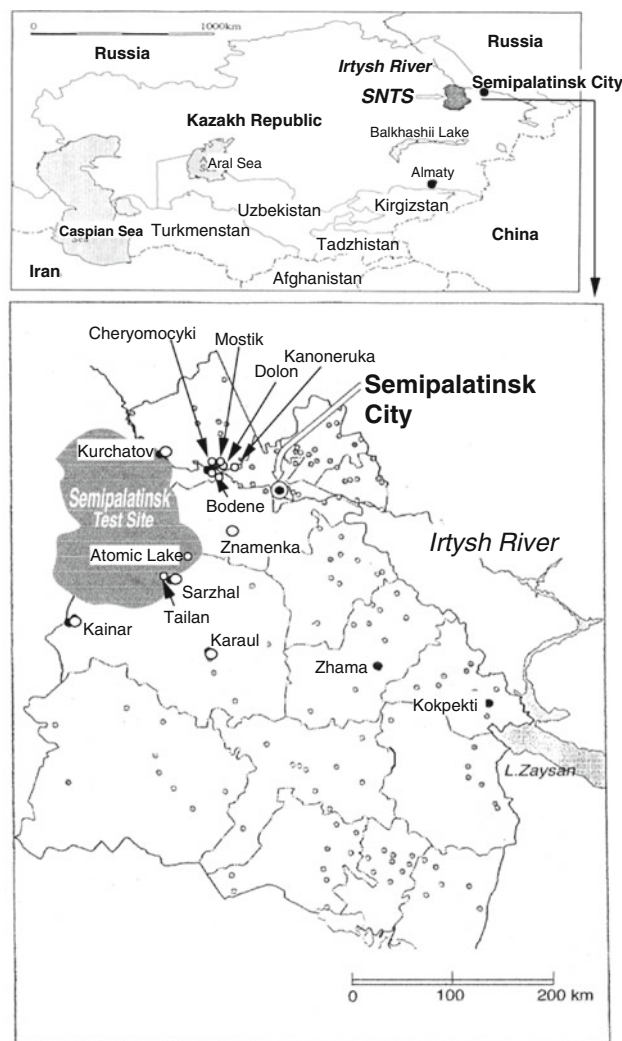


Fig. 1 Map showing sampling locations of well waters around Semipalatinsk Nuclear Test Site, Kazakhstan

Measurement of uranium

Uranium isotopes were determined by α -particle spectrometry after radiochemical separation [11]. The sample water was at first acidified to less than pH 1 by adding a small amount of HNO_3 . After shaking and standing for overnight, the water was transferred into a 500 mL beaker with the addition of known amount of ^{232}U as a yield tracer, and evaporated to dryness. The obtained residue was dissolved in 10 M HCl and the solution was passed through an anion-exchange resin column (Dowex 1 \times 8 of 100–200 mesh, Cl^- form, $0.8\text{ cm}^{\phi} \times 5\text{ cm}$). The column was washed with a small amount of 8 M HNO_3 to remove adsorbed iron and then by a sufficient amount of 10 M HCl to remove most of the other elements. Uranium was eluted from the column with 2 M HCl and the solution was evaporated to dryness. The separated U was electroplated

onto a polished stainless steel disc (2 cm^φ), and its activity was measured by α -particle spectrometer with measuring time of 3–4 days (Tennelec TC256 spectrometer coupled to a 1k-channel pulse height analyzer). The counting efficiency is about 30% and the lowest limit of detection is about 0.2 mBq for ²³⁸U.

Analysis of major chemical compositions

Major dissolved ions (Na⁺, K⁺, Mg²⁺, Ca²⁺, Cl⁻, NO₃⁻ and SO₄²⁻) of water samples were determined by ion chromatograph (Dionex ICS-1000). The alkalinity was measured by titration method with 0.1 M HCl down to pH 4.8 [12].

Results and discussion

Chemical composition

The chemical composition of 35 investigated waters is listed in Table 1. The pH of water samples ranges from 7.0 to 8.1, and is mostly neutral. The cation/anion balance ($(\Sigma_{\text{cation}} - \Sigma_{\text{anion}})/(\Sigma_{\text{cation}} + \Sigma_{\text{anion}})$ in meq/L) for most of samples measured is smaller than 5%, although some samples have values over 10%. Total dissolved salt (TDS) concentrations are in the range 189–936 mg/L. As a whole, the TDS seems to be higher in Dolon, Mostik, Budene, Znamenka, Salzhal and Karaul than in other settlements. Those water samples contain large amounts of Na⁺, Ca²⁺, Mg²⁺, HCO₃⁻ and SO₄²⁻.

Uranium levels and isotopic ratios

The result of the uranium analysis of well water samples is summarized in Table 2, together with those of river water samples from the Irtysh River. It is apparent from Table 2 that concentrations of ²³⁸U in the investigated waters vary in a wide range 3.6–356 mBq/L (0.3–28.7 $\mu\text{g/L}$), with a factor of about 100. The lowest concentration (3.6 mBq/L) of ²³⁸U was detected in the well water from Kainar and the highest concentration (356 mBq/L) of ²³⁸U was observed in the well water samples from Karaul. In Dolon and Karaul, the ²³⁸U contents change widely even within the area of each settlement. For waters from other settlements, the concentrations of ²³⁸U do not change largely. Such variation of ²³⁸U concentrations may be connected with the different local, geological and hydrological conditions of the original places of the investigated waters, although the ²³⁸U levels seem to increase with increasing mineralization as a whole. Literature values of ²³⁸U in freshwater have been reported in the range 0.002–5 $\mu\text{g/L}$, and the median was 0.4 $\mu\text{g/L}$ [13]. Global means, 0.04 and 2.0 $\mu\text{g/L}$, were

reported [14]. The ²³⁸U contents found here are several to several tens of times higher than the reported values. Kawabata et al. [15, 16] have also observed similarly high ²³⁸U concentrations for well water samples collected from some areas in Kazakhstan and Uzbekistan. It is worth noting that the ²³⁸U concentrations in some well samples from Dolon, Tailan, Sarzhal and Karaul settlements are comparable to or higher than the World Health Organization's restrictive proposed guideline of 15 $\mu\text{g (U)/L}$ [17]. Most of the measured ²³⁴U/²³⁸U activity ratios are higher than 1, and vary from 1.5 to 7.9, with most from 1.5 to 3. Those higher ratios of ²³⁴U/²³⁸U may be explained by preferential leaching of ²³⁴U due to the α -recoil effect [11, 18]. No clear relationship is observed between ²³⁸U concentration and ²³⁴U/²³⁸U activity ratio. On the other hand, all of the measured ²³⁵U/²³⁸U activity ratios show the value of around 0.046 with a relatively large counting error of about 10%. It is probable that the elevated concentration of ²³⁸U found in some settlements around the SNTS is not due to the close-in fallout from nuclear explosions at the SNTS, but rather to the intensive weathering of rocks including U there.

The ²³⁸U concentrations for river water samples collected from the Irtysh River are 31–37 mBq/L (2.5–3.0 $\mu\text{g/L}$). The levels in the Irtysh River are close to the values in well water samples from Mostik and Cheryomocyki. The ²³⁴U/²³⁸U activity ratios (1.7–1.8) are nearly the same as those found at settlements around the Irtysh River.

Radiological annual dose

Assuming that a man drinks 2 L of water per day, the annual effective doses (*D*) resulting from consumption of the investigated waters can be calculated using the following formula:

$$D (\mu\text{Sv/y}) = \sum I_i \cdot F_i \cdot 365$$

where *I_i* is the concentration of the given U isotopes (Bq/day), and geometric mean values of ²³⁸U and ²³⁴U concentrations were used as representative values for each settlement. The ²³⁵U contents were estimated by using the value (0.046) of ²³⁵U/²³⁸U activity ratio for natural uranium. The values of *F_i* are the ingestion dose coefficients (dose equivalent per intake of unit activity, Sv/Bq) reported by the International Commission on Radiological Protection [19]; 4.4×10^{-8} for ²³⁸U, 4.9×10^{-8} for ²³⁴U and 4.9×10^{-8} for ²³⁵U. In this case, fractional transfer to blood is assumed to be 0.02 for all uranium isotopes. The effective dose for adults caused by ingestion of uranium isotopes of the investigated well waters are presented in Table 3, except the settlements where only one well water sample was measured. The calculated effective doses vary

Table 1 Chemical composition of the investigated well and river waters

Settlement	Sampling date	Ion concentration (mg/L)										Ion balance (%) ^a
		pH	Na	K	Mg	Ca	Cl	NO ₃	SO ₄	HCO ₃	TDS	
Kanoneruka												
00Y71	08.11.00	8.1	4.77	n.d.	6.7	53.23	51.1	0.75	38.1	73	228	−0.4
Dolon												
05D1	09.20.05	7.7	93.6	19.3	24.2	62.7	33.9	71.2	114	260	679	5.1
05D2	09.20.05	7.4	120	4.0	28.4	76.7	48.6	114	132	293	817	3.2
04D1	11.11.04	7.6	92.7	4.6	21.8	66.0	39.7	55.7	116	273	670	1.7
04D2	11.11.04	7.2	116	3.7	26.9	81.7	66.0	104	138	236	773	5.2
04D3	11.11.04	7.2	93.1	17.3	22.9	61.3	46.5	70.2	119	269	699	0.5
04D4	11.11.04	7.4	81.5	2.7	17.5	51.0	28.5	48.8	97.1	242	569	0.0
04D5	11.11.04	7.5	73.6	4.0	16.7	49.7	19.9	10.3	65.2	301	541	0.9
00D1	08.11.00	7.4	61.5	n.d.	23.2	107	100	11.5	129	266	699	−0.8
00D2	08.11.00	7.4	134	n.d.	30.9	121	94.1	15.5	253	294	942	5.2
Mostik												
Y69	08.12.04	7.8	14.3	n.d.	17.4	122	95.2	17.4	62.4	199	528	4.2
Cheryomocyki												
03CH1	10.20.03	7.4	14.3	0.3	10.8	21.2	7.1	5.0	17.2	113	189	1.8
03CH2	10.20.03	7.5	37.0	29.5	21.5	38.3	34.7	25.6	48.2	129	363	14.6
03CH3	10.20.03	7.5	20.8	6.3	18.9	37.2	15.9	17.1	29.5	117	262	15.8
03CH4	10.20.03	7.6	29.1	6.5	12.1	22.9	11.1	5.3	21.3	121	229	11.8
Bodene												
03BW1	10.22.03	7.5	201	1.9	61.1	47.6	111	39.5	211	174	847	19.0
03BW2	10.22.03	7.9	231	1.3	38.0	30.0	121	13.0	261	180	874	10.1
03BW3	10.22.03	8.1	219	1.4	38.0	19.1	109	7.2	280	176	851	6.8
Znamenka												
05Z1	09.22.05	7.0	111	1.4	37.5	41.7	50.4	106	130	208	686	4.1
Tailan												
W-7	10.10.99	n.a.	226	8.8	35.7	130	118	n.d.	399	n.a.		
Sarzhai												
05S1	09.22.05	7.6	114	2.4	29.9	70.3	59.2	18.7	228	214	736	3.6
05S2	09.22.05	7.8	117	2.3	40.7	92.9	84.6	65.5	222	271	896	2.4
99S1	10.10.99	n.a.	131	n.d.	40.8	142	102	3.5	252	n.a.		
99S2	10.10.99	n.a.	102	10.8	35.2	119	72.5	n.d.	189	n.a.		
95S1	10.05.95	n.a.	95.5	5.3	40.1	131	104	36.2	203	n.a.		
95S2	10.05.95	n.a.	140	5.2	58.8	147	162	81.6	272	n.a.		
Kainar												
99KA1	10.11.99	n.a.	16.3	4.6	7.1	73.7	10.5	n.d.	50.4	n.a.		
99KA2	10.11.99	n.a.	25.6	10.4	10.7	87.8	25.4	2.4	53.1	n.a.		
Karaul												
04K1	11.12.04	7.9	84.3	6.1	37.6	132	84.2	177	198	216	936	2.4
04K2	11.12.04	7.7	62.9	2.3	31.4	110	44.6	69.0	185	262	767	1.5
04K3	11.12.04	8.0	23.7	2.1	16.2	76.4	9.1	7.9	111	203	450	1.6
04K4	11.12.04	7.2	35.7	3.2	22.3	101	28.1	47.1	109	223	569	6.5
04K5	11.12.04	7.5	30.8	2.6	20.1	86.0	16.6	18.6	114	242	531	1.7
04K6	11.12.04	7.9	23.7	2.1	16.1	76.2	9.2	8.0	112	210	457	0.5
99K1	10.10.99	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.	n.a.		
Irtysh River												
05R1	09.20.05	8.0	20.0	1.5	7.3	31.9	21.2	1.3	20.7	140	244	−4.4

Table 1 continued

Settlement	Sampling date	Ion concentration (mg/L)										Ion balance (%) ^a
		pH	Na	K	Mg	Ca	Cl	NO ₃	SO ₄	HCO ₃	TDS	
04R1	08.12.04	8.3	11.6	n.d.	8.2	58.9	24.7	n.d.	52.0	110	266	6.9

Error shows 1 σ standard deviation from counting statistics

^a $(\Sigma \text{cation} - \Sigma \text{anion})/(\Sigma \text{cation} + \Sigma \text{anion}) \times 100$

n.d. not detected, *n.a.* not analyzed

Table 2 Results of uranium measurements of the investigated well and river waters

Settlement	²³⁸ U concentration		Activity ratio	
	(mBq/L)	($\mu\text{g/L}$)	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U
Kanoneruka				
00Y71	11.4 \pm 0.7	0.92	1.63 \pm 0.12	0.047 \pm 0.008
Dolon				
05D1	189.9 \pm 6.8	15.3	1.63 \pm 0.03	0.044 \pm 0.002
05D2	157.2 \pm 8.7	12.6	1.64 \pm 0.06	0.043 \pm 0.002
04D1	99.8 \pm 5.0	8.03	1.44 \pm 0.05	
04D2	194.6 \pm 12.1	15.7	1.54 \pm 0.06	
04D3	197.9 \pm 11.4	15.9	1.48 \pm 0.05	
04D4	45.1 \pm 1.4	3.62	1.65 \pm 0.05	
04D5	51.4 \pm 2.8	4.13	1.51 \pm 0.07	
00D1	135.6 \pm 6.3	10.9	1.42 \pm 0.06	0.040 \pm 0.005
00D2	117.1 \pm 5.4	9.42	1.36 \pm 0.06	0.048 \pm 0.004
Mostik				
Y69	49.2 \pm 2.2	3.96	1.54 \pm 0.07	0.050 \pm 0.010
Cheryomocyki				
03CH1	42.3 \pm 1.9	3.40	1.36 \pm 0.06	0.047 \pm 0.006
03CH2	25.1 \pm 0.7	2.02	1.44 \pm 0.05	
03CH3	22.6 \pm 0.7	1.81	1.38 \pm 0.05	0.043 \pm 0.006
03CH4	28.9 \pm 0.7	2.33	1.53 \pm 0.04	
Bodene				
03BW1	144.3 \pm 4.8	11.6	2.93 \pm 0.06	0.048 \pm 0.005
03BW2	114.2 \pm 4.9	9.19	2.98 \pm 0.08	0.043 \pm 0.005
03BW3	139.9 \pm 5.2	11.2	2.67 \pm 0.06	0.049 \pm 0.006
Znamenka				
05Z1	36.5 \pm 2.5	2.94	6.80 \pm 0.38	0.045 \pm 0.005
Tailan				
W-7	274.0 \pm 10.9	22.0	1.97 \pm 0.06	0.049 \pm 0.003
Sarzhall				
05S1	144.4 \pm 6.1	11.6	2.42 \pm 0.06	0.044 \pm 0.003
05S2	213.9 \pm 12.0	17.2	2.41 \pm 0.07	0.043 \pm 0.003
99S1	206.4 \pm 14.1	16.6	2.35 \pm 0.13	0.048 \pm 0.003
99S2	127.8 \pm 6.9	10.3	2.19 \pm 0.11	0.052 \pm 0.006
95S1	156.1 \pm 8.3	12.6	2.15 \pm 0.10	0.050 \pm 0.006
95S2	172.4 \pm 10.0	13.9	2.26 \pm 0.12	0.045 \pm 0.005
Kainar				
99KA1	3.56 \pm 0.59	0.29	7.88 \pm 1.28	
99KA2	4.14 \pm 0.67	0.33	5.51 \pm 0.87	

Table 2 continued

Settlement	²³⁸ U concentration		Activity ratio	
	(mBq/L)	($\mu\text{g/L}$)	²³⁴ U/ ²³⁸ U	²³⁵ U/ ²³⁸ U
Karaul				
04K1	82.8 \pm 4.9	6.66	2.34 \pm 0.10	
04K2	90.8 \pm 6.6	7.30	2.16 \pm 0.11	0.048 \pm 0.005
04K3	52.1 \pm 3.6	4.19	2.53 \pm 0.14	
04K4	116.8 \pm 7.4	9.39	1.51 \pm 0.07	0.045 \pm 0.004
04K5	97.6 \pm 5.1	7.85	1.73 \pm 0.07	0.048 \pm 0.005
04K6	48.6 \pm 2.8	3.91	2.35 \pm 0.11	
99K1	355.6 \pm 23.4	28.6	1.09 \pm 0.06	0.047 \pm 0.009
Irtys River				
05R1	31.0 \pm 1.3	2.49	1.77 \pm 0.07	0.044 \pm 0.005
04R1	37.0 \pm 1.5	2.98	1.65 \pm 0.06	

Error shows 1 σ standard deviation from counting statistics

from 1.0 to 18.7 $\mu\text{Sv/y}$. The dose (18.7 $\mu\text{Sv/y}$) estimated for the adults living in the Sarzhall region is comparable to the value of 16 $\mu\text{Sv/y}$ (range 9–20 $\mu\text{Sv/y}$) reported recently by Vintró et al. [20]. Those doses are lower than WHO and IAEA reference value (100 $\mu\text{Sv/y}$) for drinking water [21, 22].

Conclusion

The concentrations of ²³⁸U in the well water samples from some settlements around the SNTS vary in a wide range 3.6–356 mBq/L (0.3–28.7 $\mu\text{g/L}$). The ²³⁸U concentrations in some samples from Dolon, Tailan, Sarzhall and Karaul settlements are comparable to or higher than the World Health Organization's restrictive proposed guideline of 15 μg (U)/L. The measured ²³⁴U/²³⁸U activity ratios are higher than 1, and are mostly from 1.5 to 3. The ²³⁵U/²³⁸U activity ratios show the value of around 0.046, indicating that U in the wells is of natural origin. It is probable that the higher ²³⁸U concentrations are not due to the close-in fallout from nuclear explosions at the SNTS, but rather to the intensive weathering of rocks including U there. The calculated annual effective doses arising from the ingestion

Table 3 Annual effective dose to adult arising from U ingestion (consuming 2 L of water daily) through well water in each settlement

Settlement	Analyzed number of samples	Range	Geometric mean			Effective dose (mSv/y)
			^{238}U (mBq/L)	^{234}U (mBq/L)	$^{235}\text{U}^a$ (mBq/L)	
Dolon	9	45.1–197.9	117.3	177.9	5.4	10.3
Cheryomocyki	4	22.6–42.3	28.9	41.1	1.3	2.4
Bodene	3	114.2–144.3	132.1	377.4	6.1	18.0
Sarzhai	6	127.8–213.9	167.3	365.4	7.7	18.7
Kainar	2	3.56–4.14	3.8	25.3	0.2	1.0
Karaul	7	48.6–355.6	94.6	181.9	4.4	9.8

^a Contents of ^{235}U were calculated by using the value (0.046) of $^{235}\text{U}/^{238}\text{U}$ for natural U

of U isotopes (^{234}U , ^{235}U and ^{238}U) are in the range 1.0–18.7 $\mu\text{Sv/y}$, and are lower than the recommended value of 100 $\mu\text{Sv/y}$ for drinking water.

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